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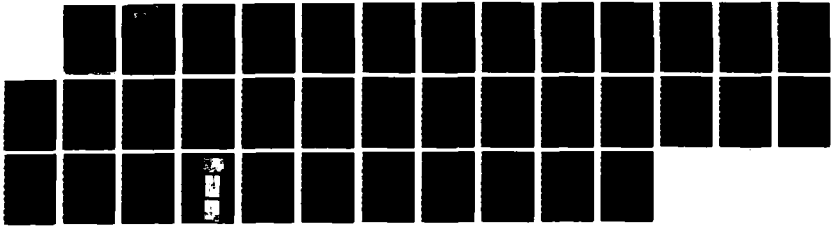

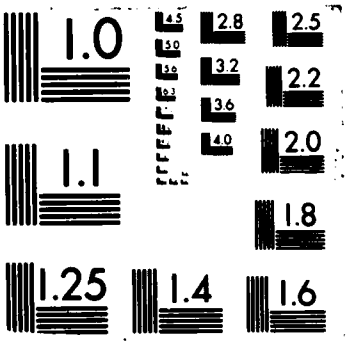
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MULTIPHOTON IONIZATION OF ATOMS *

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ABSTRACT

Studies of multiphoton ionization of atoms have revealed several unexpected characteristics. The confluence of the experimental evidence leads to the hypothesis that the basic character of the atomic response involves highly organized, coherent motions of entire atomic shells. The important regime, for which the radiative field strength E is greater than an atomic unit (e/a_o^2), can be viewed in approximate correspondence with the physics of fast ($\sim$ 10 MeV/amu) atom-atom scattering. This physical picture provides a basis for the expectation that stimulated emission in the x-ray range can be produced by direct highly nonlinear coupling of ultraviolet radiation to atoms.

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The invention of the laser made possible the experimental study of the nonlinear interaction of radiation with matter. And, subsequently, over the last two decades, a considerable field of activity has developed around that basic problem, which can be represented generically by the process

$$N\gamma + X \rightarrow X^* \qquad (1)$$

in which N photons lead to the excitation of a target atom or molecule X. For generality, the excited product X* can denote either bound or continuum motions of the constituent particles. This article will deal with certain recent findings concerning multiphoton ionization. The context of this physical study, however, relates directly to another problem of general fundamental significance and one that has had an important influence on the direction and purpose of the work at the University of Illinois at Chicago. That problem concerns the development of a laser at x-ray wavelengths, a long-sought goal.

Historically, the initial discussions of coherent generation in the x-ray range (1) and nonlinear atomic emission (2) and absorption (3) all appeared more than twenty years ago in entirely independent circumstances. Recently, however, these two areas of inquiry have become strongly linked, and it now appears that the achievement of the former may depend, at least in one possible representation, on certain basic properties of the latter.

The significance of the x-ray laser goal is easily stated. A spectrally bright source of radiation in the x-ray region would be unsurpassed (4,5) in its ability to microvisualize condensed matter. There is little doubt that major areas of application would include basic materials research, microelectronics, biology, and, indeed, any field that requires structural information of solid matter on an atomic scale. Since the peak spectral brightness of an x-ray laser is expected to be on the order of one trillion times greater

than any alternative means, it would be an ideal source of radiation for these purposes. At the same time, it is undisputed that matter, under appropriate conditions, can provide the amplification in the x-ray range ($\hbar\omega \sim 10^3 - 10^4$ eV) necessary to construct a coherent source. Unfortunately the achievement of those suitable conditions has proven to be a formidable task.

The difficulty in generating amplification at a quantum energy of 1 KeV is apparent from the general requirement, established by basic physical reasoning, that extraordinarily high specific power densities, on the order of 10^{14} W/cm^3 or greater, be applied in a carefully controlled way (1,6,7). How can these necessary conditions best be produced? It is generally understood (1,6,7) that (i) a high effective brightness source of excitation and (ii) an appropriate physical coupling mechanism are the key requirements for the successful creation of the conditions for amplification in the x-ray range. Although several alternative approaches are currently being evaluated, over the last few years our work has centered on the use of (i) high-power ultraviolet lasers to serve as the source of excitation and (ii) nonlinear radiative processes to provide the needed physical coupling. In order to evaluate whether this idea could work, we began our study of multiple quantum ionization.

A simple representation of the overall process is illustrated in Fig. 1. Obviously, if $\hbar\omega_x$ is in the kilovolt range and if $\hbar\omega$ represents an ultraviolet quantum, processes with values of N exceeding 100 must occur with appreciable probability in order for this mechanism to be useful. Therefore, a single fundamental question emerges: what are the basic physical principles that establish the limit on N?

When we began to study this question, no relevant experimental data on this matter existed. This was particularly true for radiative field strengths

in the vicinity of an atomic unit, (e/a_o^2). Extant theoretical work, however, predicted ridiculously low rates for high-N processes and, if believed, would automatically lead to the conclusion that any proposal to utilize a mechanism, such as that shown in Fig. 1, is preposterous. These earlier theories, however, were based on rather restrictive assumptions concerning the basic nature of the electronic motions governing the nonlinear amplitudes. On the other hand, several years ago I found it possible to construct a class of high-order processes, involving certain types of atomic motions, in which the rates would be enormously enhanced. Are such motions possible? I now believe that the method used to calculate those original estimates was in many ways incorrect, although the conclusion derived may not be. In any case, the basic question had been raised and resolution of the issue could only come from experiment.

EXPERIMENTAL STUDIES

Basically, there are three categories of fundamental physical measurement founded, respectively, on the spectroscopy of (i) ions, (ii) electrons, or (iii) photons, that can be used to unravel the nature of the physical processes involved in the nonlinear interactions under study. In our work we have used ion charge state spectra, photoelectron energy spectra, and the properties of scattered radiation. Among these, the measurement of ion charge state spectra under collision-free conditions provides a simple, unambiguous experimental test that gives direct information on the scale of the energy transfer rate between the radiation field and the atom. Therefore, as a first step, such experiments were performed. The results of these initial studies were surprising, for it was from measurements (8,9) of this kind that the first suggestions of an anomalously strong coupling for high-order processes

($N \sim 100$) were obtained.

The general class of physical processes studied in those experiments examining ion charge state spectra (X^{q+}) was

$$N\gamma + X \rightarrow X^{q+} + qe^{-}. \qquad (2)$$

Some of the apparatus used in the work described below is illustrated in Fig. 2, which shows the 193-nm ultraviolet laser system, the time-of-flight ion spectrometer, and the time-of-flight electron analyzer. Available ultraviolet laser technology makes the performance of such studies convenient, since extraordinarily high brightness and, therefore, unusually large focused intensities are possible with these laser sources. Initially, studies (8) of the process represented by Eq. 2, a subclass of the general reaction in Eq. 1, were conducted with 193-nm radiation at an intensity of $\sim 10^{14}$ W/cm^2.

Fig. 3 illustrates the range of intensity that has been explored over the past two years along with a projection of the experimental regime that will become available over a comparable period in the future. Currently, peak intensities in the range of 10^{16} to 10^{17} W/cm^2 can be produced with pulses having a duration of a few picoseconds at a wavelength of 193 nm. At this intensity, the electric field E is comparable to an atomic unit $E_o = e/a_o^2$. Furthermore, as shown in Fig. 3, it is believed that technical advances in femtosecond ultraviolet laser technology in the near future will enable peak intensities, for coherent energy, to be produced in the range of 10^{20} to 10^{21} W/cm^2. This would represent a radiation field amplitude of $\sim 100\ E_o$ and, in terms of energy density, would be equivalent to that produced by a black body with a temperature close to 10 keV, an environment characteristic of thermonuclear sources. Although all the experiments to date have been conducted in the range of $E \lesssim E_o$, considerable attention in the discussion below will be given to this interesting and important regime for which $E >> E_o$.

A prominent feature of the studies of the ionic charge spectra was the unusually strong nonlinear coupling found characteristic of certain heavy materials (8-11), a feature apparent even at intensities in the vicinity of 10^{14} W/cm^2. These experiments clearly demonstrated that standard theoretical techniques were incapable, by a discrepancy as great as several orders of magnitude, of describing the observed charge state spectra. Interestingly, a resemblance was perceived between the observed ionic charge state spectra and those known to be characteristic of Auger cascades (9-12). In addition, subsequen work by us, as well as other studies (14-15) conducted at the wavelengths of 1.06 and 0.53 μm, confirmed the anomalous nature of the coupling strength. There was no doubt that the findings of these experiments were clearly in contradiction to all theoretical treatments, of which there is a considerable number (16-21). This unexpected result, of course, stimulated further experiment

In experiments to determine the role of atomic structure on the coupling mechanism, the ionic spectra of several elements from He (Z = 2) to U (Z = 92) were studied (8-11). A typical ionic spectrum for Xe, produced by 193-nm radiation at an intensity of $\sim 10^{16}$ W/cm^2 with pulses of $\sim$ 5 ps duration, is illustrated in Fig. 4. One immediately notes the presence of all charge states up to Xe^{8+}, the first five of which are seen to have approximately comparable abundances. An overall summary of the ionic species registered in the survey of the atomic number dependence is presented in Fig. 5. As shown in Fig. 5, the maximum observed energy transfers are on a scale of several hundred electron volts for certain heavy materials.

One of the salient features of the data is the apparent influence of atomic shell structure (8) on the observed ion spectra. This dependence is manifested prominently in the behavior of the heavier rare gases. For Ar, Kr, and Xe, the maximum charge states observed correspond to the complete removal of certain outer atomic sub-shells. Indeed, for these materials they are the 3p,

the 4p, and both the 5s and 5p shells, respectively.

The hint provided by the role of the shell structure led to the hypothesis that it was mainly the number of electrons in the outer subshells that governed the coupling. A measurement of the response of elements in the lanthanide region, with the use of a method involving laser-induced evaporation (to provide the material, enabled this view to be checked. As one moves from La (Z = 57) to Yb (Z = 70) in the lanthanide sequence, aside from slight rearrangements (23) involving the 5d shell for Gd (Z = 64), 4f electrons are being added to interior regions (24) of the atoms. The data illustrated in Fig. 5 for Eu ($4f^7 6s^2$) and Yb ($4f^{14} 6s^2$), which differ by seven 4f electrons, indicate that these inner electrons play a small role in the direct radiative coupling, a fact that is in rapport with the observed dependence on the outer-shell structure.

The intensity dependence of these ion spectra was also examined (10,11). Over the range of intensities studied ($\sim 10^{15}$ - 10^{17} W/cm^2), higher intensity generally translated into an increased yield of ions of a particular charge, although not necessarily an increase in the maximum charge state observed. For example, the ion Xe^{8+}, with closed shell ground state (25) configuration $4d^{10}$, was the greatest charge state detected at $\sim 10^{16}$ W/cm^2, and, although its abundance increased at $\sim 10^{17}$ W/cm^2, no Xe^{9+} appeared at the higher intensity. This observation has led to the interpretation that the 5p and 5s electrons in Xe are the ones that govern the direct coupling of the atom to the ultraviolet radiation field.

It was also possible to obtain information on the frequency dependence of the coupling by comparing the results at 193 nm (10) with studies performed independently at 1.06 μm and 0.52 μm (14,15). This comparison,

which was conducted for an intensity of $\sim 10^{14}$ W/cm^2, for both Kr and Xe, indicated that the energy transfer rate was reduced at the longer wavelengths.

Overall, the ion studies showed (i) anomalously strong nonlinear coupling for certain heavy materials, (ii) an unmistakable signature of atomic shell effects, and (iii) that energy transfer rates were generally greater at shorter wavelengths of irradiation. Moreover, the experimental evidence strongly suggested, at least in a first approximation, that the greater the number of electrons in the outer shell, as designated solely by the principal quantum number, the greater the strength of the nonlinear coupling (8-11).

Since it is expected that the measurement of photoelectron energy distributions could provide valuable information on the detailed nature of the electronic motions occurring in reactions such as that shown in Eq. 1, experiments of that type, performed under collision-free conditions, have been conducted (26). This expectation, indeed, appears to be borne out. For example, substantial differences in the electron distributions produced by Ar and Kr were seen even though the ion spectra for these materials are similar and show that the outer p-shell is completely stripped in both cases (10,11).

The most significant results now available, however, appear in connection with the behavior of Xe. Indeed, in contrast to Ar and Kr, the Xe electron energy spectrum exhibits a dramatic change with increasing intensity of the 193-nm radiation in the range of 10^{14} to 10^{15} W/cm^2. The first ionization line, which corresponds to two-photon absorption with a corresponding photoelectron energy of 0.7 eV, nearly disappears, while the three-photon process, arising from continuum-continuum transitions (27,28), becomes

dominant. Furthermore, the final state distribution of the ions generated for the three-quantum process has approximately 80 percent in the excited $5s^2 5p^5\ ^2P_{1/2}$ state, with the remaining 20 percent in the $5s^2 5p^5\ ^2P_{3/2}$ ground level.

In addition to the ladder of continuum-continuum lines, new sharp photoelectron features appear in the range from 8 to 22 eV at an intensity of $\sim 10^{15}$ W/cm^2. These lines have been assigned to $N_{45}00$-Auger lines after excitation of the 4d inner-shell. The most prominent lines are those associated with $N_{45}0_1 0_1$ transitions, which terminate on the $4d^{10}5s^0 5p^6$ double-hole state. This identification is predicated on the observation that their relative spacing, number, and, to a somewhat lesser extent, relative intensities fit well to values previously reported (29,30) for such Auger transitions. Moreover, a total of eight electron lines is observed, representing a quartet of pairs of transitions, all of which exhibit the known (29,30) $4d_{3/2}$ - $4d_{5/2}$ splitting in xenon of $\sim$ 2 eV.

The general trend (31,32) of the electron spectrum for xenon as a function of intensity is illustrated in Fig. 6. Note the appearance of a group of lines at an intensity of $\sim 10^{15}$ W/cm^2 which are attributed to Auger decay of 4d-vacancies in the atom. This spectral region (26), which is believed to represent $N_{45}00$ Auger processes, is shown in higher resolution in Fig. 7.

Finally, it should be stated that further experimental evidence bearing on the mechanism of coupling is present in the characteristics of certain stimulated emission spectra that have been observed in krypton (33-36). In this case, the states believed to be involved are those having multiple excitations and inner-shell excitations (37-41) in closely coupled subshells,

such as $4s4p^6n\ell$ and $4s^24p^4n\ell n'\ell'$. As the discussion in Section III shows, this class of levels is of exactly the type expected to be strongly excited if certain highly organized atomic motions, which are consistent with both the ion charge state and photoelectron spectra, are driven by the radiation field.

MECHANISM OF COUPLING

We are now in a position to interpret the experimental findings in terms of a specific, although highly speculative, model for the atomic response. The main purpose of the description given below is not to provide an exact definitive analysis, which now obviously is impossible, but rather to furnish a framework for the process of physical reasoning that will reveal the true nature of these very nonlinear mechanisms.

From an analysis of the data now at hand, which includes information on the dependencies on the atomic number (8-10) (Z), the intensity, the frequency, and the polarization, the following hypothesis has emerged as an approximate description of the basic character of the electronic motions involved in these processes. Overall, the data strongly indicate that an organized motion of an entire shell, or a major fraction thereof, is directly involved in the nonlinear coupling. With this picture, the outer atomic subshells are envisaged as being transiently driven in coherent oscillation by the intense ultraviolet wave. With this type of electronic motion, the observed increase in the multiphoton coupling strength can be qualitatively correlated (10) with the larger magnitude of the effective charge involved in the interaction. In quantum mechanical language, an oscillating shell would be represented by a wave function of a multiply excited configuration. In this way, a multielectron atom undergoing a nonlinear interaction responds

in a fundamentally different fashion from that of a single electron counterpart. Interestingly, a class of multiply excited configurations, consisting of doubly excited states, is known (42) to play an important role in processes of single quantum photoionization. With this interpretation, the results of our studies of multiquantum ionization simply indicate a nonlinear analog of this basic electronic mechanism.

In principle, the response of an atom to a pulsed external field with an amplitude approaching an atomic unit ($E \sim e/a_o^2$), if calculated with full rigor in the framework of a time-dependent many-body theory, would present the possibility of a nearly unbounded level of complication. Therefore, in order to advance our understanding of this problem, a simpler approximate form of analysis has to be found. An appropriately formulated treatment, which correctly represents the basic nature of electronic motions, however, should be able to describe qualitatively the principal characteristics of the experimental observations. These include (i) the basic coupling strength and resulting energy transfer rate, (ii) the shell effects, (iii) the origin of the strong nonlinearity, (iv) the frequency characteristics, and (v) the ability to produce atomic inner-shell excitation.

In order to achieve that goal, a relatively simple model (10,11), valid at sufficiently high intensity, can be contemplated. In this case, we imagine an atom composed of two parts: an outer shell of electrons (a) driven in coherent oscillation by the radiative field and a remaining atomic core (b) for which direct coupling to the radiation field is neglected. In this picture, coupling between these two systems can occur, since the outer electrons could, through inelastic "collisions," transfer energy to the core. Simple estimates (11) indicate that, for intensities corresponding to an electric field $E >> e/a_o^2$, enormous oscillating atomic

current densities j on the scale of 10^{14} to 10^{15} amps/cm^2 could be temporarily established in the outer regions of the atom. For ultraviolet radiation under these conditions, the electrons in the outer atomic shell can be accelerated to mean kinetic energies considerably higher than $\sim$ 10 KeV, a value far above their respective binding energies (11). Furthermore, in the limit of high intensity, it is possible to formulate an estimate of the coupling of the coherently driven outer electrons with the remaining atomic core by relatively simple procedures. This is now done at two levels of approximation, initially with the neglect of the influence of the coherence characterizing the motion of the outer electrons and, subsequently, with its inclusion.

An estimate can now be furnished based simply on the magnitude of the ambient current density j. Since the electron kinetic energies are considerably above their corresponding binding energies, it is possible to use a first-order Born approximation (43) in a manner similar to that used to study electron collisions for K- and L-shell ionization (44) and shell-specific ionization processes in highly charged ions (45,46). Indeed, in the case of xenon ions, actual measured cross sections for electron impact ionization are available (47).

In this elementary classical picture (11), the transition rate R can be written as

$$R \simeq \frac{j}{e} \sigma_e \qquad (3)$$

in which e is the electronic charge and σ_e is the cross-section characterizing the excitation of the atomic core by inelastic electron collisions arising from the current density j. If $j \tilde{=} 10^{14}$ A/cm^2 and $\sigma_e \tilde{=} 10^{-19}$ cm^2, then $R \tilde{=} 6 \times 10^{13}$ sec^{-1}. Furthermore, if the radiatively driven current

density j is damped by electron emission in a time τ on the order of $\sim 10^{-15}$ seconds, an approximate time scale characterizing autoionization, the overall transition probability $P \gtrsim R\tau \gtrsim 6 \times 10^{-2}$, indicating a significant probability of energy transfer.

The characterization of the outer-shell motion as a simple current density j, however, fails to take into account the fact that the electronic motions are generated through interaction with a coherent wave. It is anticipated that the coherence associated with the motions of the outer-shell electrons induced by intense irradiation will have important consequences (48) for the coupling of energy to atomic inner-shells that were ignored in the simple estimate given above. In order to describe the influence of this type of coherent atomic motion, it is now related to certain properties (49) of energetic ($\gtrsim$ 10 MeV/amu) atom-atom collisions.

The role of coherence in the motion of the outer electrons in the excitation of the core is readily described in terms of energetic atom-atom (A/B) collisions. In this comparison (48), a correspondence is established, as shown in Fig. 8, between the scattering of the coherently driven outer electrons (a) from the atomic core (b) and the respective interaction of the electrons in the projectile atom A with the target atom B. Consider the process

$$A + B(o) \overset{\sigma_{no}}{\rightarrow} A + B^*(n) \qquad (4)$$

in which A is a ground-state neutral atom with atomic number Z_A and B*(n) represents an electronically excited configuration of the target system with quantum numbers collectively represented by (n). In the plane-wave Born approximation (PWBA), the cross-section $\sigma_{r.o}$ can be written in the form presented by Briggs and Taulbjerg (49) as

$$\sigma_{no} = \frac{8\pi e^4}{v^2} \int_{K_{min}}^{K_{max}} |\varepsilon_{no}^{B}(\vec{K})|^2 [|Z_A - \sum_j \omega_j \langle \phi_j^A | \exp(i\vec{K}\cdot\vec{s}_A) | \phi_j^A \rangle|]^2 \frac{dK}{K^3} \qquad (5)$$

in which

$$\varepsilon_{no}^{B}(\vec{K}) = \int dr_B^3 \psi_{nB}^{*}(\vec{r}_B) \exp(i\vec{K}\cdot\vec{r}_B)\psi_{oB}(\vec{r}_B). \quad (6)$$

In Eqs. 5 and 6, e is the electron charge, Z_A is the atomic number of the projectile atom, v is the relative atom-atom velocity, ϕ_j^A are orthonormal spin-orbitals representing the electrons on the projectile atom with spatial coordinate s_A, ω_j is the statistical weight of the shell, $\vec{K}$ is the momentum transfer in the collision, and ψ_{oB} and ψ_{nB} represent the electron wave functions of the target system as a function of the spatial variable r_B. The summation over the index j appearing in Eq. 5 extends over all occupied orbitals so that, in the limit $\vec{K} \to 0$, the summation tends to the number of electrons N_A associated with the projectile atom (49). In the low-momentum transfer limit, in which complete screening occurs, the amplitudes of the electrons combine coherently, and the contribution to the cross-section σ_{on} arising from the motion of the electrons in atom A is increased by a factor of N_A^2 over that of a single electron at the same collision velocity v. Equivalently, for sufficiently low momentum transfer such that $Ka_o \ll \hbar$, the electron cloud acts as a coherent scattering center with a mass $N_A m_e$, a charge $N_A e = Z_A e$, a velocity v, and a kinetic energy $N_A(1/2\ m_e v^2)$. Significantly, because of the coherence, the single particle energies $(1/2\ m_e v^2)$ add so that, in principle, this value could be below the magnitude required to produce the excitation of the target atom B.

In sufficiently high field strengths, it appears that coherently accelerated electrons in outer atomic shells (a) can interact with the remaining atomic core system (b) in a manner closely analogous to the atom-atom scattering described above. If a PWBA description is used, the cross-section representing energy transfer can be written by inspection from Eq. 5 with $Z_A = 0$. The basic physical concepts are simply represented in the

high field limit ($E >> e/a_o^2$), a regime in which the driven electronic velocities correspond approximately to those characteristic of atom-atom collisions at a collision energy of $\sim$ 10 MeV/amu. Therefore, the motion of these electrons can simulate the electronic collisional environment that would occur in fast atom-atom encounters, but with the important absence of the nuclear contribution arising from the Z_A term in Eq. 5.

In order to see how this mechanism would scale with the basic physical parameters involved, the contribution σ_{on} can now be estimated for inner-shell excitation arising from coherently excited atomic shells. For this Eq. 5 is taken with $Z_A = 0$ and K_{max} restricted to $\lesssim \hbar/a_o$ to fulfill the condition for complete shielding. Further, Z_1 can be taken to denote the number of electrons in the outer shells and Eq. 6 expanded for $\varepsilon_{no}^B(\vec{K})$ in the customary fashion so that only the leading dipole term x_{on} is retained. Finally, for a core excitation energy ΔE we put $K_{min} \simeq \Delta E/v$, the condition that holds for ΔE much less than the collision energy. With these modifications, the full coherent piece σ_{on}^c can be written as

$$\sigma_{on}^c \simeq \frac{8\pi e^4 x_{on}^2 Z_1^2}{v^2\hbar^2} \int_{\Delta E/v}^{\hbar/a_o} \frac{dK}{K}, \tag{7}$$

a result which, with the exception of the restriction on K_{max} and the Z_1^2 factor, is exactly the form of the well-known result for inelastic scattering of electrons on atoms developed by Bethe (50). The final result, valid for

$$\alpha\left(\frac{v}{c}\right)\left(\frac{m_e c^2}{\Delta E}\right) > 1 \tag{8}$$

is

$$\sigma_{on}^c \simeq 8\pi\alpha^2\left(\frac{c}{v}\right)^2 Z_1^2 x_{on}^2 \ln\left[\alpha\left(\frac{v}{c}\right)\left(\frac{m_e c^2}{\Delta E}\right)\right] \tag{9}$$

in which α is the fine-structure constant.

Obviously, all types of possible excited configurations cannot fully benefit from this type of coherent motion, regardless of the field strengths used. Indeed, the limitation can be estimated from Eq. 8. At sufficiently high intensity in the limit $v \to c$, the maximum value of ΔE_{max} is given by

$$\Delta E_{max} \sim \alpha m_e c^2 = 3.73 \text{ KeV}. \tag{10}$$

The physical picture presented above also enables a statement concerning the frequency of irradiation ω to be formulated. For the excitation of inner-shell states in the kilovolt range by the quasi-free coherently driven motion of outer-shell electrons, two basic assumptions are involved. The first, as noted above, concerns the field strength E such that the condition

$$E >> E_o = e/a_o^2 \tag{11}$$

holds, enabling the electrons to be regarded as approximately free. The second consideration involves the energy scale of the motion, ε_e, in this case taken to be sufficiently great to excite readily inner-shell states in the desired kilovolt range. With the neglect of relativistic corrections, the electron energy can be expressed as

$$\varepsilon_e = 1/2\ m_e v_e^2 \tag{12}$$

with the quantity v_e representing the velocity of induced electronic motion. For a free electron, the maximum value of v_e, commonly known as the quiver velocity, is given by

$$v_e = \frac{eE}{m_e \omega} \tag{13}$$

for a field with angular frequency ω (51).

For stated values of E and ε_e that fulfill the assumptions of the model, a frequency scale generally characteristic of those physical conditions is now defined by combination of Eqs. 11, 12, and 13. If we take $E \tilde{=} 3E_o$ to satisfy Eq. 11 and $\varepsilon_e \sim 10^3$ eV as reasonable values, then

$$\omega \simeq (30)\alpha^3 \frac{c}{\lambda\!\!\!^{-}_c} \tag{14}$$

a frequency that corresponds to an ultraviolet wavelength of $\sim$ 200 nm. With this result, we are led to the interesting conclusion that ultraviolet wavelengths naturally match the physical conditions characteristic of the coherent atomic motions envisaged in this description. Experimental results are in agreement with this conclusion, since, as shown by the ion charge state studies described above, the observed energy transfer rates for infrared and visible radiation were reduced with respect to those characteristi of the ultraviolet range.

This simple model can also be used to estimate the threshold condition for 4d-vacancy production in xenon. Although these preliminary results (26) do not constitute a proof of the mechanism involved, it is simply observed that energy transfer from coherently driven valence shell electrons could produce such inner-shell excitation. Furthermore, with the model presented above, along with consideration of the known (47) inelastic electron scattering cross-sections for xenon ions, an estimate can be made of the intensity at which such Auger lines should appear. Inelastic scattering studies (47) show that the 4d excitation in xenon has a threshold at $\sim$ 67.6 eV, closely followed by a broad maximum at $\sim$ 100 eV. If the motion of the N_A outer electrons in xenon is approximated as that of free electrons, the maximum electronic kinetic energy ε_e can be written, in a form which reexpresses Eq. 12, as

$$\varepsilon_e = (1.79 \times 10^{-13}) \lambda^2 I \qquad (15)$$

with units of electron volts for ε_e, micrometers for λ, and Watts per square centimeter for I. The 4d-threshold at $\sim$ 67.6 eV corresponds to an intensity I for single electron motion of $I \sim 10^{16}$ W/cm^2, a value somewhat above that used in the actual experimental studies (26,31) for photoelectron production. However, if the picture of the coherent motion is valid, the single-particle

energy can be reduced, for a fixed threshold requirement, by a factor of Z_1, the number of electrons participating in the coherent outer-shell motion. For xenon, previous ion studies (8,10,11), data from which are shown in Fig. 4, indicated that $Z_1 = 8$ is a reasonable value, which is the total number of electrons in the n = 5 shell ($5s^2 5p^6$). This reduces the threshold intensity for 4d-vacancy production to $\sim 1.2 \times 10^{15}$ W/cm^2, a value quite close to that ($\sim 10^{15}$ W/cm^2) corresponding to the experimental appearance of the electron lines presumed to arise from Auger decay shown in Figs. 6 and 7.

CONCLUSIONS

Basic physical studies of collision-free nonlinear atomic processes, through an analysis involving combined measurements of ion charge state distributions, photoelectron energy spectra, and photon spectra arising from intense ultraviolet irradiation, have produced data that strongly indicate that multielectron atoms respond in a manner fundamentally different from single-electron counterparts. The confluence of the evidence suggests that, under appropriate circumstances, the outer atomic subshells can be driven in coherent oscillation, and this ordered electronic motion can, by direct intra-atomic coupling, lead to the rapid excitation of atomic inner-shell states. Quantum mechanically, such states of motion for the outer-shell electrons would be described by multiply excited configurations. Two direct consequences of this type of motion are (i) that the maximum magnitude of the oscillating intra-atomic electric field can approach several atomic units, since the fields of all the participating outer electrons combine constructively, and (ii) that the harmonic content of the resulting field can, because of the nonlinear character of the electron-electron $1/r^2$ coulombic interaction, become large. A strong, highly nonlinear

intershell coupling results, and enhanced rates of nonlinear absorption are expected.

An elementary atomic model, formulated to take advantage of certain simplifications that appear to be characteristic of the high-intensity regime ($E >> e/a_o^2$), has enabled qualitative comparisons to be made among several of the most prominent experimentally observed properties. Although this representation is only at the hypothetical stage, the five points of contact are (i) the basic coupling strength, (ii) the shell effects, (iii) the origin of the strong nonlinearity, (iv) the frequency characteristics, and (v) the ability to produce atomic inner-shell excitation. This general, although approximate, form of analysis has additional value, since it enables us to estimate the response of atoms throughout the periodic table and, thereby, provides a set of testable hypotheses for comparison with future experiments. It is important to add that, since the original preparation of this article, results obtained by much more quantitative calculations (52) involving the time-dependent Hartree-Fock method basically confirm (i) the fundamental character of atomic motion, represented as the analogy with fast atom-atom collisions, even for field strengths $E \sim E_o = e/a_o^2$ (32,48), and (ii) provide remarkable quantitative agreement as well. For the latter, the intensity levels at which 4d Auger electrons should be observable in xenon correspond, within a factor of approximately two, for both theoretical approaches. Importantly, these results are also in accord with experimental figures (26), to within the same rather small level of uncertainty.

An atom in a radiative field whose amplitude is significantly greater than an atomic unit experiences a violent perturbation that has important features in common with certain well-studied collisional phenomena, such as ion-atom collisions, electron-ion collisions, and beam-foil interactions (53,

Indeed, in the case of beam-foil collisions, a radiative environment at an intensity of 3×10^{18} W/cm^2 and an ultraviolet wavelength approximates (32), in several important respects, the conditions associated with the passage of an argon ion through a carbon foil with a kinetic energy of $\sim$ 1 GeV. This similarity leads to the concept (55) of an "optical solid," in which stationary atoms in a sufficiently intense radiative field will experience an interaction comparable to that of energetic ions traversing solid matter. The consequence is an extreme level of excitation on the scale required to establish the conditions needed to produce stimulated emission in the kilovolt range. In addition, the coherence of the radiative environment can act to introduce a measure of control on the energy transfer that will enable considerable selectivity in the energy flow to be achieved. If this speculative hypothesis survives, a synthesis of many areas of atomic physics and an unexpected nexus between the original research concerning coherent x-ray production and nonlinear processes will result.

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FIGURE CAPTIONS

Fig. 1: Simple representation of an atomic multiquantum process involving the absorption of N ultraviolet quanta ($\hbar\omega$) to produce an appropriate upper state X* which is inverted with respect to the population in a lower level X_ℓ. Since a kilovolt energy scale is assumed for $\hbar\omega_x$, N >> 1 and X* and X_ℓ will generally lie far above the ionization energy of the neutral atomic ground state system X.

Fig. 2: Pictures of experimental apparatus used in the performance of the studies of multiphoton ionization at the University of Illinois at Chicago. (a) Output amplifier (left) of the gigawatt 193 nm picosecond laser system; (b) ion time-of-flight spectrometer; (c) electron time-of-flight spectromenter.

Fig. 3: Representation of the present and future ranges of intensity of irradiation available with high brightness ultraviolet laser technology. Initial observations of anomalous processes were made at $\sim 10^{14}$ W/cm^2. Intensities comparable to thermonuclear environments ($\sim 10^{21}$ W/cm^2) appear possible with pulse lengths in the 100 fs range containing $\sim$ 1J of energy.

Fig. 4: Collision-free ion time-of-flight spectrum of xenon produced by 193 nm irradiation at an intensity of $\sim 10^{16}$ W/cm^2 with pulses having a duration of $\sim$ 5 ps. The distortion of the charge state peaks is caused by the naturally occurring isotopic distribution of xenon. The charge states of the Xe^{+q} species observed are indicated.

Fig. 5: Data concerning the multiple ionization of atoms produced by irradiation at 193 nm. Plot of total ionization energies of the observed charge states as a function of atomic number (Z). The coincidence of an H_2O^+ background signal prevented the I^{7+} species from being positively identified.

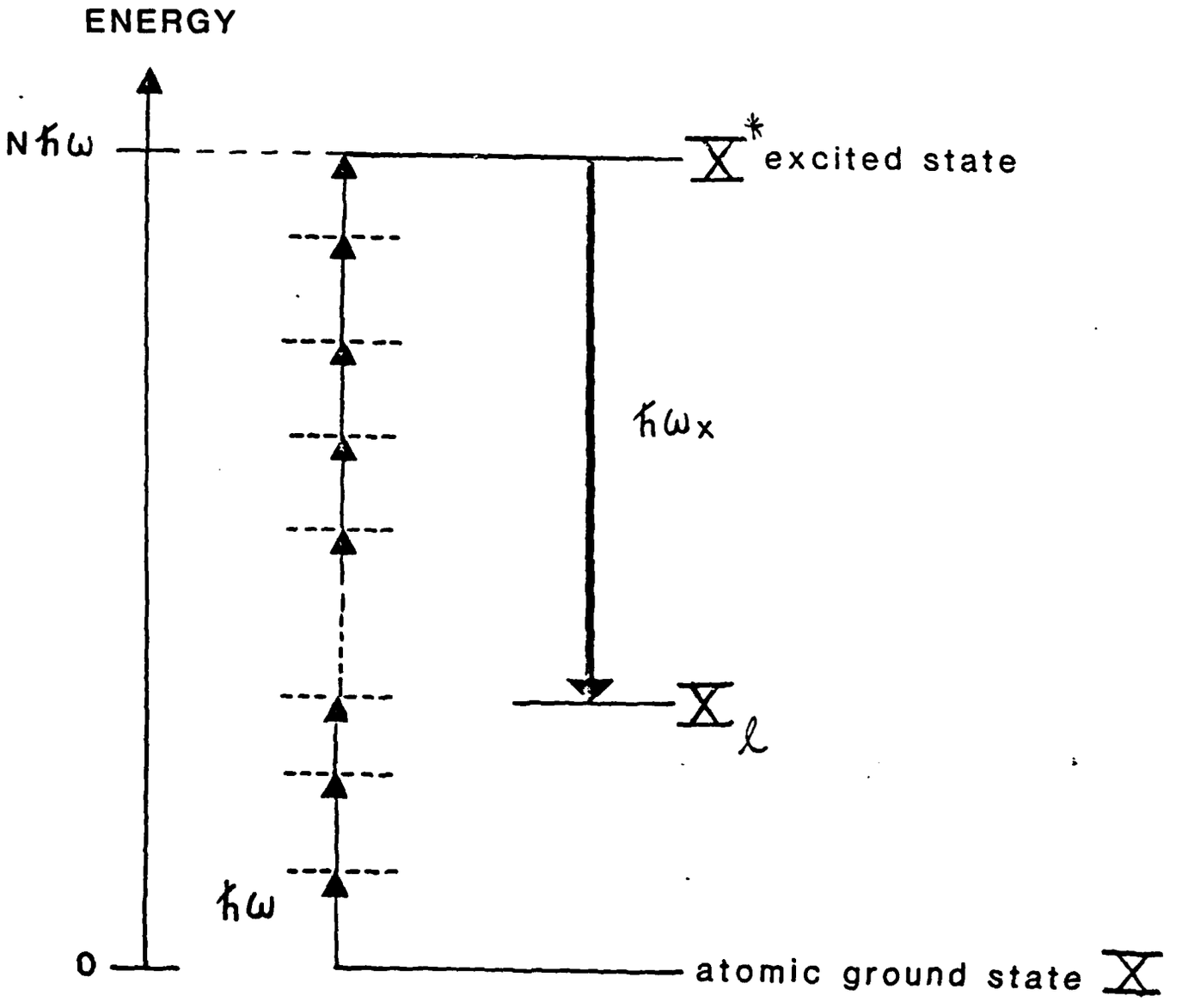
Fig. 6: Overall xenon time-of-flight photoelectron spectrum from $\sim$ 0.1 to $\sim$ 100 eV. The uncertainty in the intensity scale is approximately a factor of two. Irradiation was at 193 nm with a pulse duration of $\sim$ 5 ps with a lens with a focal distance of 20.5 cm. Electron lines originating from Xe and Xe^+ arising from two (2γ), three (3γ) and (4γ) processes are indicated along with a group tentatively assigned as Auger features.

Fig. 7: Prominent transitions observed in the electron spectrum of xenon irradiated with 193 nm at $\sim 10^{15}$ W/cm^2 are shown. Both continuum-continuum ($4\gamma \rightarrow Xe^+_{3/2}$, $Xe^+_{1/2}$) and tentatively assigned Auger ($N_{45}OO$) features are apparent. The splittings between the three N_4-N_5 pairs, two of which are shown by horizontal arrows, have the common value of $\sim$ 2 eV, the known $4d_{5/2}$-$4d_{3/2}$ separation in xenon. The vertical arrows indicate the high energy edges of the observed features which represent the true energies of the lines.

FIGURE CAPTIONS (cont.)

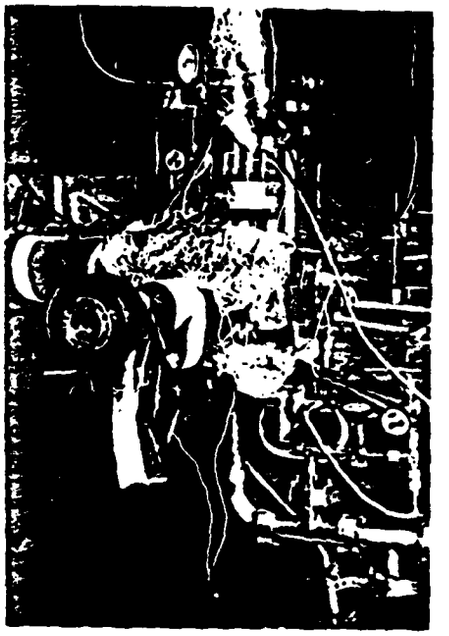
Fig. 8: Approximate correspondence between (α) A/B atom-atom collision at relative velocity v and (β) coherent relative motion of outer-shell electrons (a) with respect to core (b). For simplicity, the electrons in inset (β) are depicted as undistorted mutually displaced charge distributions moving with relative velocity v. The nuclear charges of the projectile and target systems, Z_A and Z_B, respectively, are denoted in the atom-atom collisions.

Fig 1.



(a)

(b)



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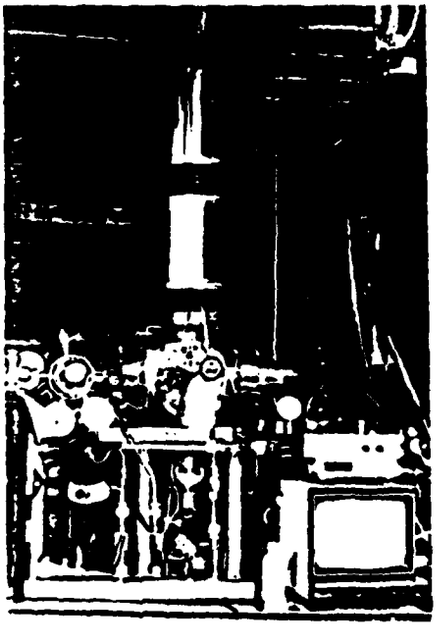
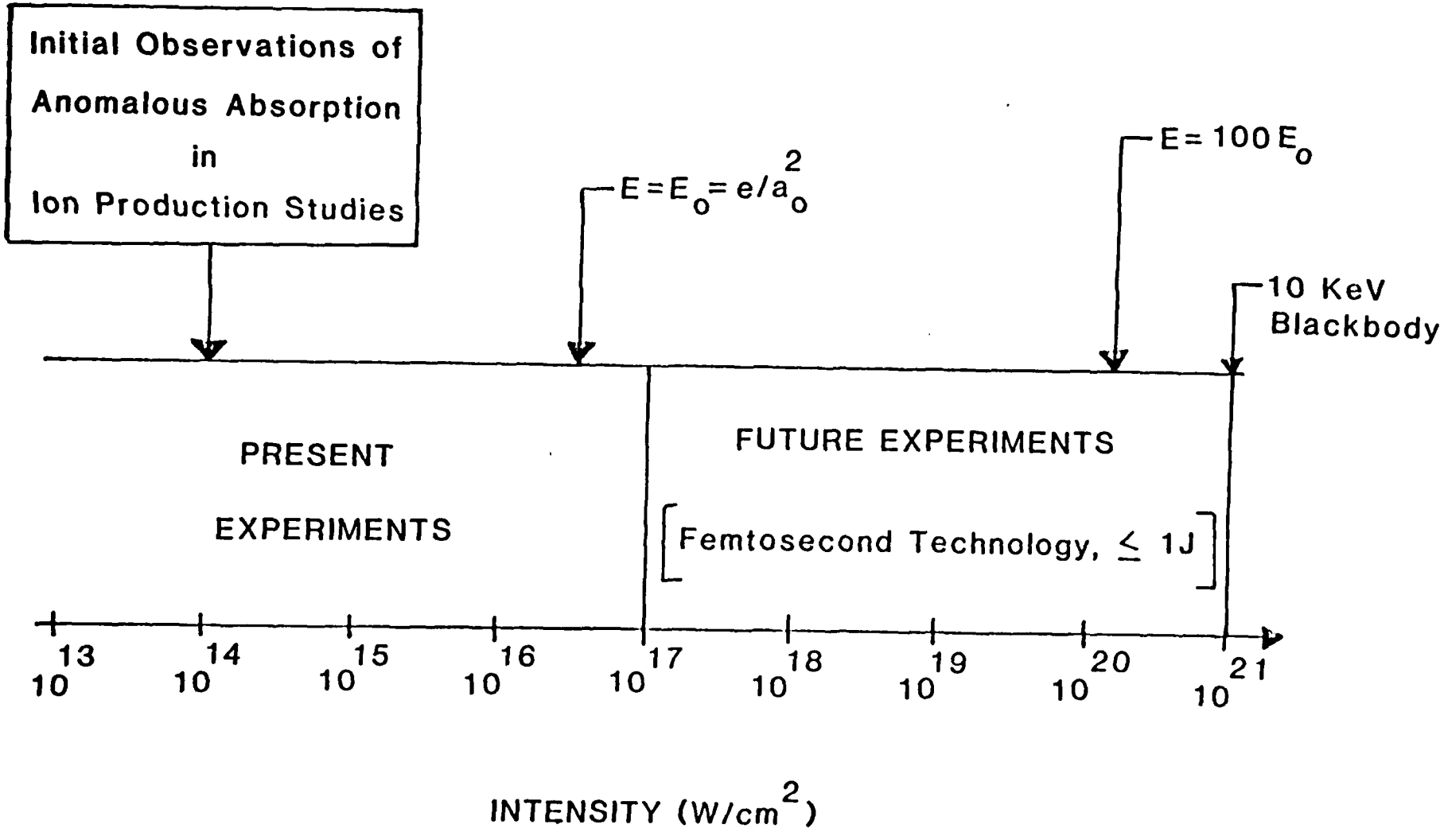
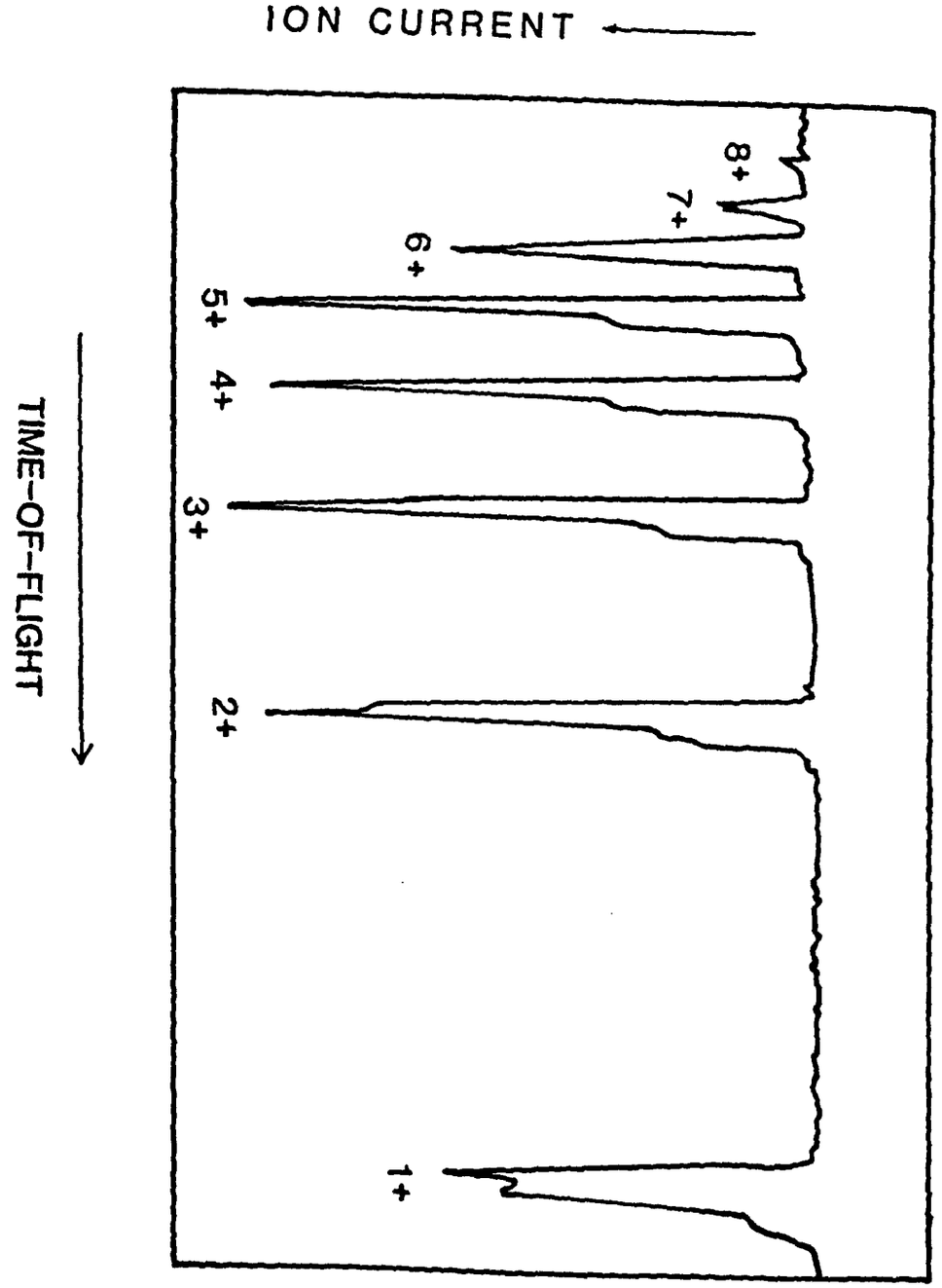


EXPERIMENTAL RANGE OF IRRADIATION



Fig(1)



Fig(5)

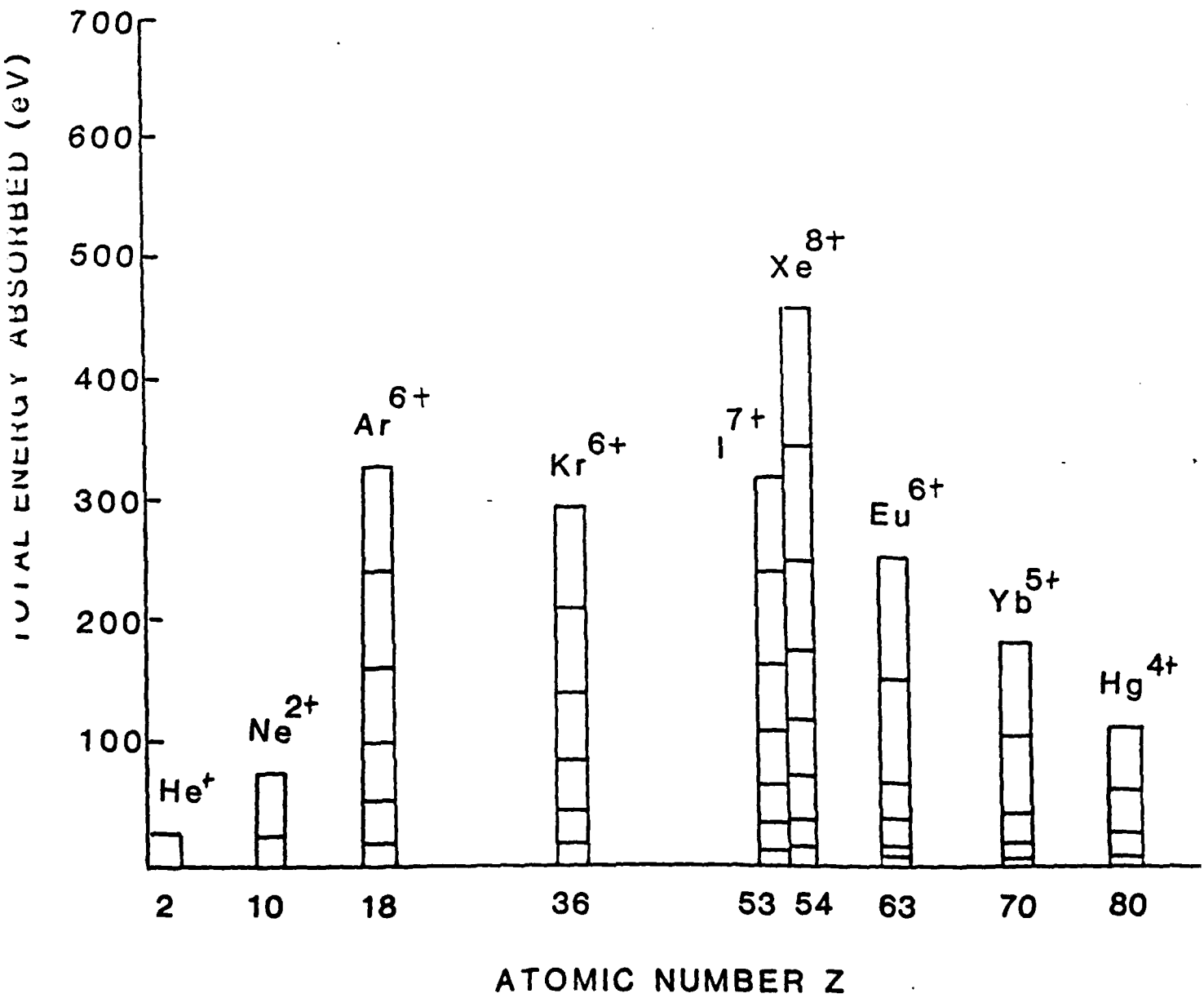

TOTAL ENERGY ABSORBED (eV)
700
600
500
400
300
200
100
He+
Ne2+
Ar6+
Kr6+
I7+
Xe8+
Eu6+
Yb5+
Hg4+
2
10
18
36
53
54
63
70
80
ATOMIC NUMBER Z

Fig (6).

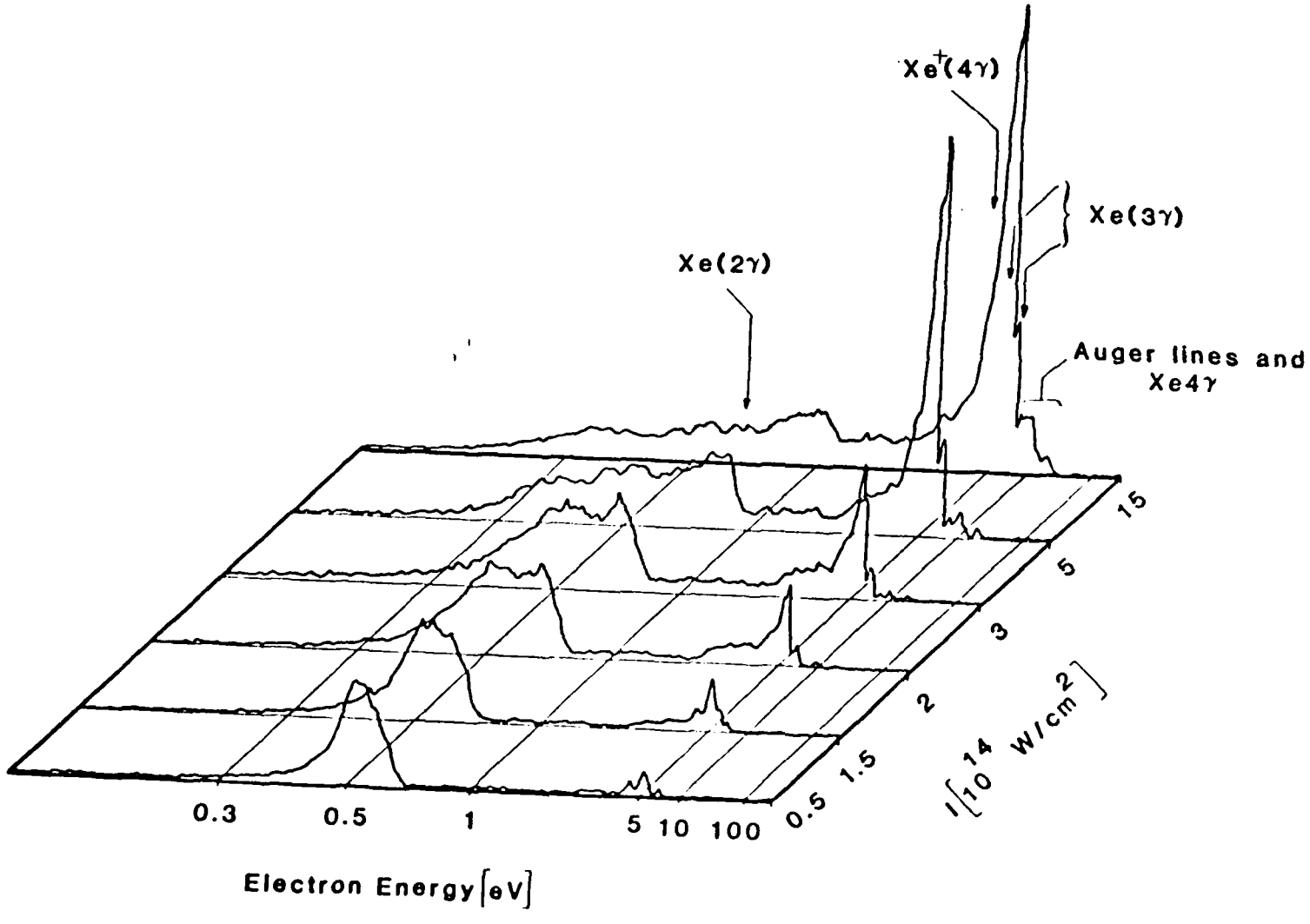

Xe+(4γ)
Xe(3γ)
Xe(2γ)
Auger lines and
Xe4γ
0.3
0.5
1
5
10
100
Electron Energy [eV]
0.5
1.5
2
3
5
15
I [10^14 W/cm^2]

Fig (7)

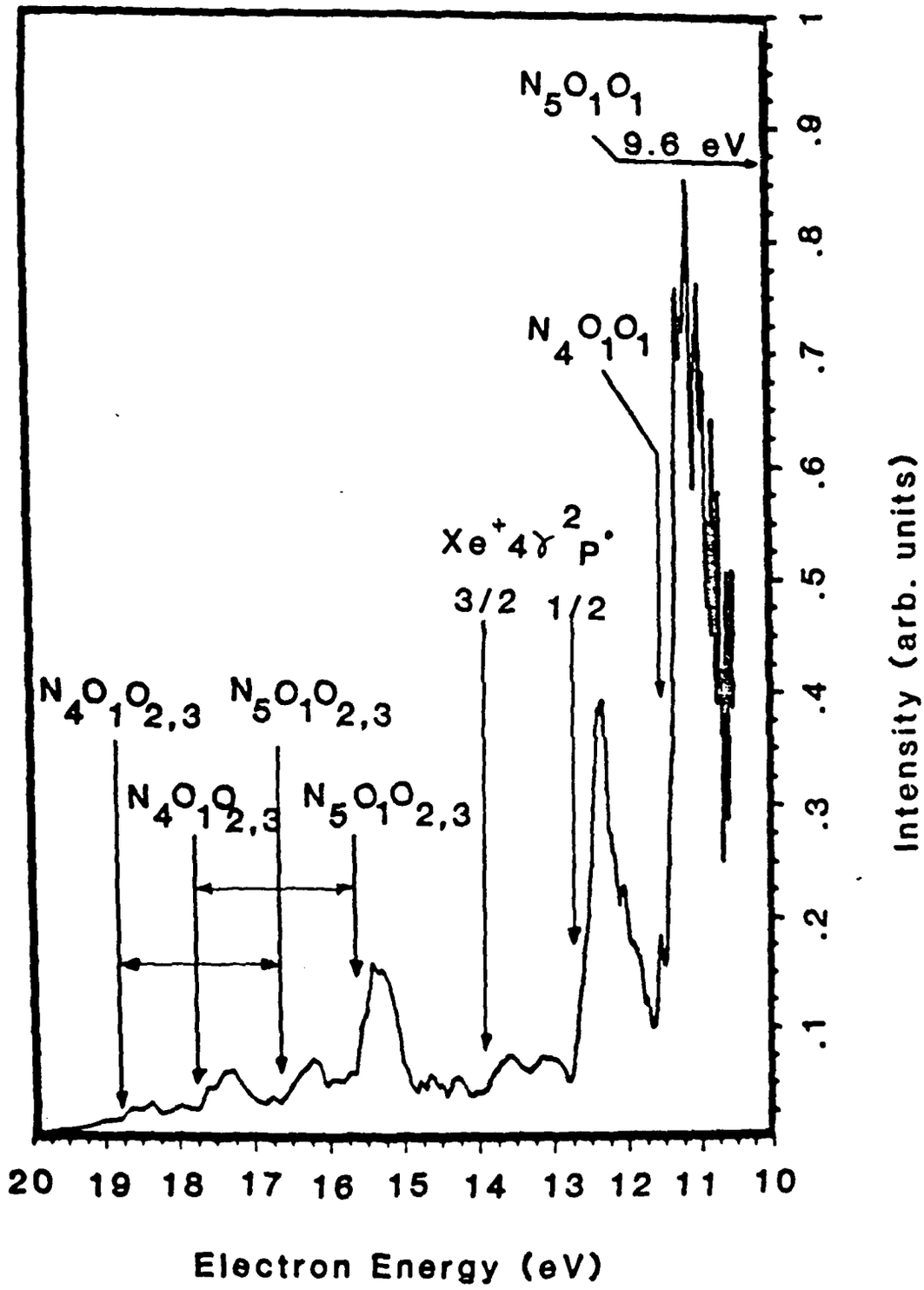
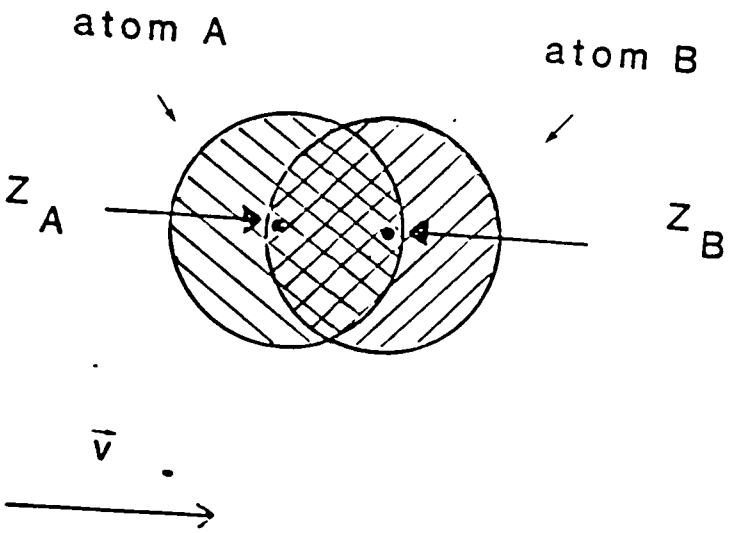
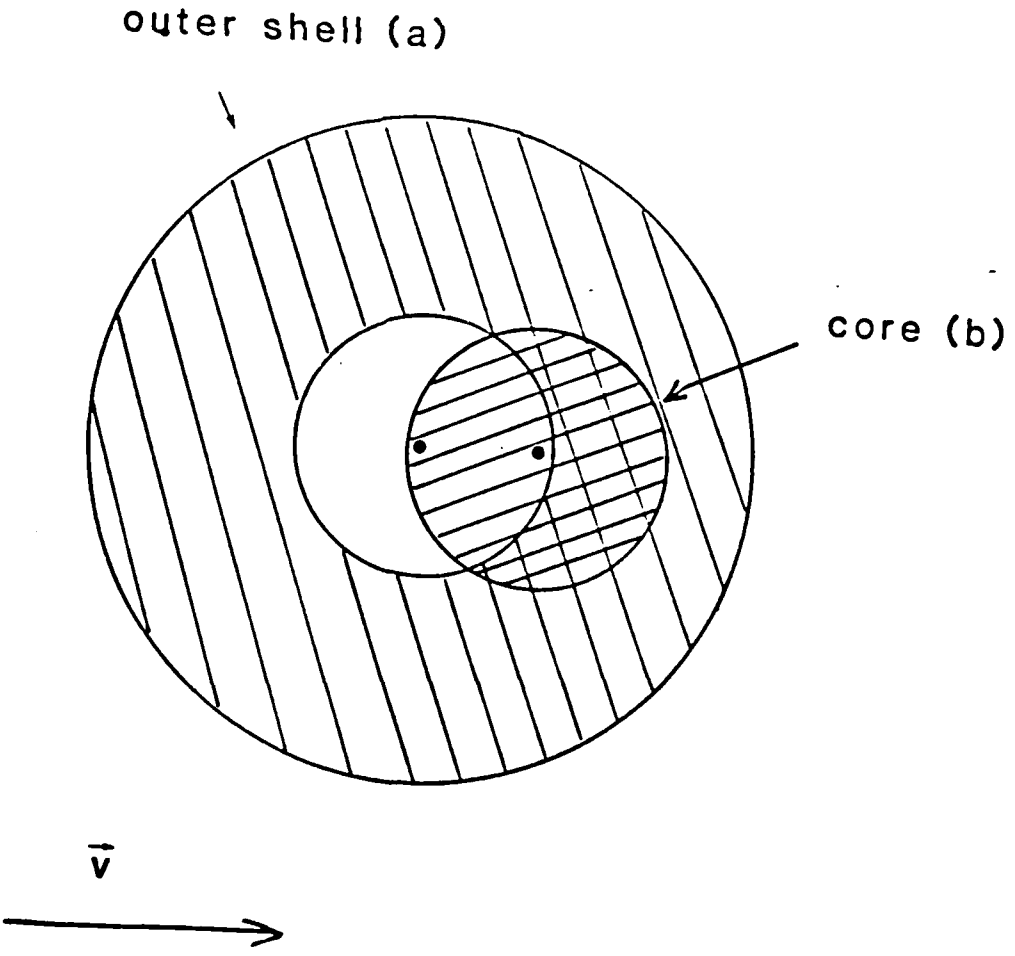

$N_5O_1O_1$
9.6 eV
$N_4O_1O_1$
$Xe^+4\gamma\ ^2P^\circ$
3/2
1/2
$N_4O_1O_{2,3}$
$N_5O_1O_{2,3}$
$N_4O_1O_{2,3}$
$N_5O_1O_{2,3}$
20 19 18 17 16 15 14 13 12 11 10
Electron Energy (eV)
Intensity (arb. units)
.1 .2 .3 .4 .5 .6 .7 .8 .9 1

Fig (8)

α)



β)



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